Infrared Breakout Session – NSLS-II Workshop
Brookhaven National Laboratory
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The IR breakout session was attended by more than 30 scientists, with diverse interests ranging from microchemical analysis of biological tissues to electronic properties of exotic superconductors. The intent of this session was to review (and update) scientific requirements, to inform the user community of the NSLS-II project proposal and how it addresses those requirements, and to continue the process of refining project details. The session consisted of a mixture of invited and contributed presentations, plus extended Q&A and discussion periods.

The workshop began with an overview of the NSLS-II project by Larry Carr (NSLS) that highlighted the relevant components for infrared, and the motivation for maintaining a separate, small storage ring. Though the proposed main NSLS-II ring will operate at a 500 ma beam current, the large aperture ports necessary for long wavelength IR might be detrimental to the NSLS-II's ultra-high brightness design goal. Therefore, the NSLS plans to integrate the existing VUV/IR ring into the NSLS-II accelerator complex and optimize its operation as a low-energy (infrared) photon source. This would include higher average current (1 ampere) and higher brightness (with top-off injection), plus an upgrade to operate at the same ~ 500 MHz RF frequency as the large NSLS-II main ring. This higher frequency will provide shorter duration pulses for time-resolved applications and may allow for a coherent emission mode to produce THz pulses. Since the storage ring will be dominated by infrared activities, additional custom ports can be added to the dipole bends for extracting extremely long wavelength radiation or providing multiple/extended sources for mid-IR microscopy with array detectors. Demand for microspectroscopy continues to grow and is expected to be the dominant infrared synchrotron technique for the foreseeable future.

Dimitri Basov (Univ. of California – San Diego) surveyed the important field of correlated electron systems and the role of infrared synchrotron radiation for studying these and other novel materials. Dimitri stressed the importance of infrared spectroscopy over the widest possible spectral range (photon energies from 0.0003 to 30 eV) for advancing our understanding of these complex systems. Since high quality specimens are commonly in the form of very small crystals, the high brightness of infrared synchrotron radiation is one of its most important characteristics. In addition to doped Mott-Hubbard insulators, Dimitri identified other types of complex systems where magnetism plays an important role. This not only includes magnetic semiconductors, but also systems that have been specifically engineered, or tailored, for a particular magnetic response. The so-called "left-handed" or "negative refractive index" materials that have been developed in the last few years are an example. Dimitri also described other important synchrotron measurement techniques (e.g. ellipsometry, magnetospectroscopy, time-resolved spectroscopy) that were individually addressed in contributed presentations later in the session.

A recurring theme for synchrotron radiation is for the study of systems under extreme conditions. In many cases, such extreme states can only be achieved in small isolated regions or for brief periods of time. A prime example of this is the study of materials at extreme temperatures and pressures – such as occurs near the center of our planet. Rus Hemley (Geophysical Laboratory of the Carnegie Institution of Washington) has led a program of high-pressure research at the NSLS U2A infrared beamline. Their work has focused on the structure of minerals under conditions found deep inside the Earth, but they have also studied much "simpler" systems such as H_2O and Van der Waals compounds that show unusual phase transitions and novel molecular phases as a function of increasing pressure. Even the simplest compound, H_2, has been observed to form into new solid phases. Rus noted that the combination of infrared and x-ray methods has been crucial for achieving an understanding of these materials. High-pressure IR spectroscopy is also being used to study the technologically relevant subjects of ultra-hard materials and hydrogen storage in solids. The quest for even higher pressures and broader spectral coverage continues. Since many materials are expected to become superconductors at high pressures, access to the far-IR / THz portion of the spectrum is needed to explore their low energy electronic properties. The production of larger synthetic diamonds is one of a number of developments toward meeting this goal.

Though diffraction effects currently limit the spatial resolution for infrared vibrational spectroscopy to a few microns, the high brightness of synchrotron infrared has allowed small collections of nanomaterials to be studied. In his presentation, Laszlo Forro (Ecole Polytechnique Federale de Lausanne - EPFL) described his study of carbon nanotubes in the form of oriented ropes. Using the U10B infrared microscope, the reflectivity of a 10 micron diameter single-wall nanotube (SWNT) rope was measured using polarized synchrotron radiation, revealing a large anisotropy in the material's electrical conductivity. Indeed, the response along the SWNT direction is suggestive of a fairly good metal. Another interesting subject is electrical transport in DNA, a material reported as being insulating, semiconducting and even metallic in other recent studies. The EPFL group measured the far-infrared transmission through an oriented DNA bundle and found no evidence for conductivity in the DC limit. Laszlo also reviewed some of the earlier far IR transmission measurements through a free-standing single crystal of the high-T_C superconductor $Bi_2Sr_2CaCu_2O_{8+x}$. This study provided rather conclusive evidence that cuprates do not have a conventional "energy gap" in their excitation spectrum, which supports the notion of a "d-wave" superconducting state. Lastly, Laszlo described his group's work with Lisa Miller using vibrational microspectroscopy to study chemical changes in diseased tissues. This is one of several investigations of disease being guided by Lisa at the U10B scanning infrared microspectrometer.

The session continued with brief contributions reflecting particular interests and requirements for a future infrared synchrotron source. Source brightness across the entire IR spectral range (including the far-IR) continues to be the most important characteristic. Laszlo Mihaly (Stony Brook University) contributed a detailed description of his group's magnetospectroscopy and spin resonance program. This is a new activity for synchrotron radiation and has attracted attention from groups at the high magnetic field laboratories in Tallahassee (NHMFL) and Grenoble. Material provided by Paul Dumas (CNRS) and Gwyn Williams (JLab) reviewed the impact of synchrotron radiation for studying the interaction of molecules with metal surfaces. This subject is now the focus of a group from BNL Chemistry and Stony Brook University using the U4IR beamline. Andrei Sirenko (New Jersey Institute of Technology) intends to use synchrotron infrared radiation for ellipsometry measurements of oxide films having unusual dielectric behaviors in the far-infrared. Andrei plans to continue working the Max Planck Institute group (Christian Bernhard et al, Stuttgart) using ellipsometry and other techniques to study carbon nanotubes, semiconductor nanopillars and manganites. Reaching frequencies down to 15 cm^{-1} in a magnetic field was one measurement requirement.

A number of speakers pointed out that the pulsed character of synchrotron radiation should continue to be used for time-resolved spectroscopy. David Tanner (University of Florida) contributed some detail on this subject, pointing out that the synchrotron's broadband character is unique for a pulsed source. David also noted that better time resolution (pulse durations approaching 1 ps) would be needed to properly address dynamics problems in conventional and high-T_C superconductors, plus other electronic systems (such as nanoparticles). Efforts by the NSLS accelerator physics group have managed to bring pulse durations down to a few hundred picoseconds at modest beam currents, but instabilities associated with the RF system of the VUV/IR ring seem to prevent any further gains. Since the ring's RF would be upgraded to a higher frequency system as part of NSLS-II, significant improvements in bunch length can be anticipated. If instabilities can be controlled, significant reductions in bunch lengths may be achieved. Larry Carr noted the results from BESSY II where bunch lengths of just a few ps have been achieved, along with the production of coherent synchrotron radiation in the far IR. This "coherent THz" mode of operation may be feasible for an upgraded VUV/IR ring. Mike Martin (Lawrence Berkeley National Laboratory) expanded on this subject in a description of the proposed CIRCE project. Their model calculations for CIRCE indicate that a stable coherent mode can be achieved in combination with a sizeable average beam current. Mike pointed out that such a source would be relevant for the study of bio-systems, especially for time-resolved probes of dynamics.

Several questions about the operation of the NSLS-II IR ring were asked during the discussion periods. Some concerned details that are yet to be worked out, e.g., how is the IR ring building accessed? Participants asked about the frequency of injections for top-off mode, and whether spectra can be acquired through an injection. Based on experience, we expect most Users will want to pause acquisition during an injection. Therefore, injections should be adequately spaced to accommodate the longest likely scan period, which is presently about 20 minutes. We expect to learn more on this subject from infrared scientists at the Swiss Light Source once their IR beamline becomes operational.

Another concern was the duration of the "downtime" required for moving the existing VUV/IR ring and beamlines to the NSLS-II site, and re-establishing operations. The Users felt strongly that this should be kept to a minimum, especially since there are few alternative infrared synchrotron facilities presently in the U.S. It was suggested that the cost savings for re-using portions of the existing VUV/IR ring might be offset by a more extensive downtime.

The user community was pleased that more space will be available for beamline instrumentation. This is especially important for mid-IR spectroscopy and microscopy where a stable environment (temperature and humidity) is important. These beamlines would plan to construct experimental hutches that would provide an added layer of climate isolation and therefore improve stability. Hutches also allow for dark environments (necessary for beam alignment) and help to control acoustic noise, whose frequency components match the modulation frequency of most spectrometers.

In summary, the community of infrared users welcomed the inclusion of a dedicated ring for low energy / infrared activities within the NSLS-II project plans. This small ring would deliver improved performance (over the existing VUV/IR ring) for all infrared users, including those requiring far-IR. The expected shorter duration pulses will be of great benefit for the study of dynamics, and the possibility of a coherent mode (during special operations) is an intriguing option.